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C. W. Trainor and S. A. Mani
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Principal Investigator: Daniel W. Trainor, (617) 389-3000, Ext. 467

Scientific Officer: Director, Physics Program, Physical Sciences Division
Office of Naval Research
800 North Quincy Street
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The overall goal of this experimental program is to identify scalable techniques that efficiently convert existing high power UV lasers to lasers operating at longer wavelengths in the visible. Two nonlinear optical conversion techniques that we have considered are: stimulated Raman and parametric conversion involving the KrF laser (248 nm).

The objective of this contract is to suggest likely acceptor atoms for each technique which will thereby allow us to evaluate some of the key technical issues

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involved. These include: the production of acceptor candidates in the gas phase (typically refractory metals), the volumetric removal of the lower laser level in the stimulated Raman approach to prevent "bottle necking" and allow recycling of the atoms during the laser pulse, and the consideration of overall system efficiency and scalability to high power.

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I. INTRODUCTION

The overall goal of this combined experimental and theoretical program is to successfully and efficiently convert using scalable techniques the output of a high power KrF laser into longer wavelengths so as to vastly improve its propagation characteristics.

Since the first reported lasing of an inert gas halogen laser, a number of similar systems have demonstrated lasing characteristics. Operating at various wavelengths, with different efficiencies, a major class of electronic transition lasers came into existence. Recently, analogous mercury halide compounds showing similar formation kinetics have been shown to lase in the visible,[1,2] albeit in high temperature (~ 275°C) cells (see Figure 1). However, the most efficient laser reported to date in this group is the KrF laser operating at 248 nm. It has also produced the highest energy outputs reported utilizing e-beam pumping and e-beam controlled discharge pumping and has a demonstrated capability for being scaled to high average power. In certain applications, especially those requiring transmission through the atmosphere, its short wavelength severely limits its usefulness. This limitation in propagation at short wavelengths arises due to absorption by atmospheric ozone and to Rayleigh scattering which increases as λ^{-4} as the wavelength gets shorter. Ozone absorption is severe for wavelengths $\lesssim$ 3000 Å.

Figure 2 shows vertical transmission from a height of 3 km as a function of wavelength. Also plotted are quantum efficiency of conversion from KrF wavelengths and the total percentage transmission of converted KrF radiation. From the figure, it is apparent that to efficiently utilize KrF laser radiation, its conversion wavelength should be between 340 and 400 nm to maximize its atmospheric transmission with minimal loss from quantum yield considerations. Xenon fluoride lasers, while possessing a more attractive wavelength for propagation, have not yet demonstrated the combined efficiency and energy density comparable to KrF. Any optical conversion scheme for altering the wavelength of KrF laser radiation to the 340 to 400 nm wavelength range could have higher overall efficiency than the XeF laser performance to date if the photon conversion efficiency is > 40%. Such efficiency for conversion is a reasonable goal for the program we are discussing here. For supporting evidence, one can look over the past year at a number of milestones that have been reported relevant to the optical conversion of UV excimer lasers. With regard to overall conversion efficiency, an XeF laser has been converted, at near unit photon conversion efficiency, using barium vapor[3]. Also, ArF and KrF have

(1) Parks, J.H., Appl. Phys. Letters 31, 192 (1977)

(2) Parks, J.H., Appl. Phys. Letters 31, 297 (1977)

(3) Djeu, N. and Burnham, R., Appl. Phys. Letters 30, 473 (1977)

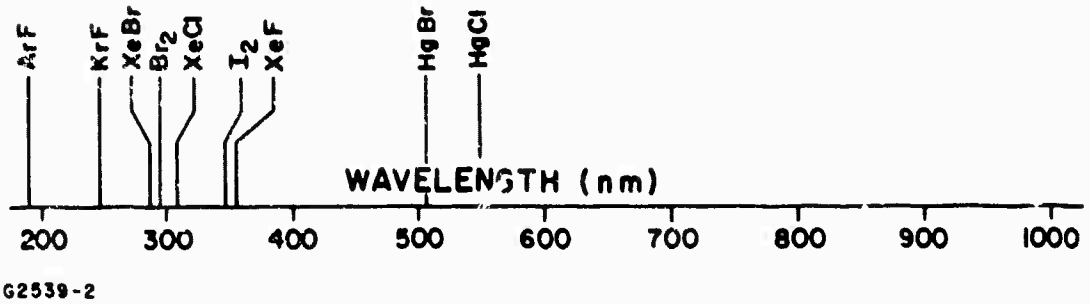

Figure 1 Demonstrated Potentially Scalable Electronic Transition Lasers

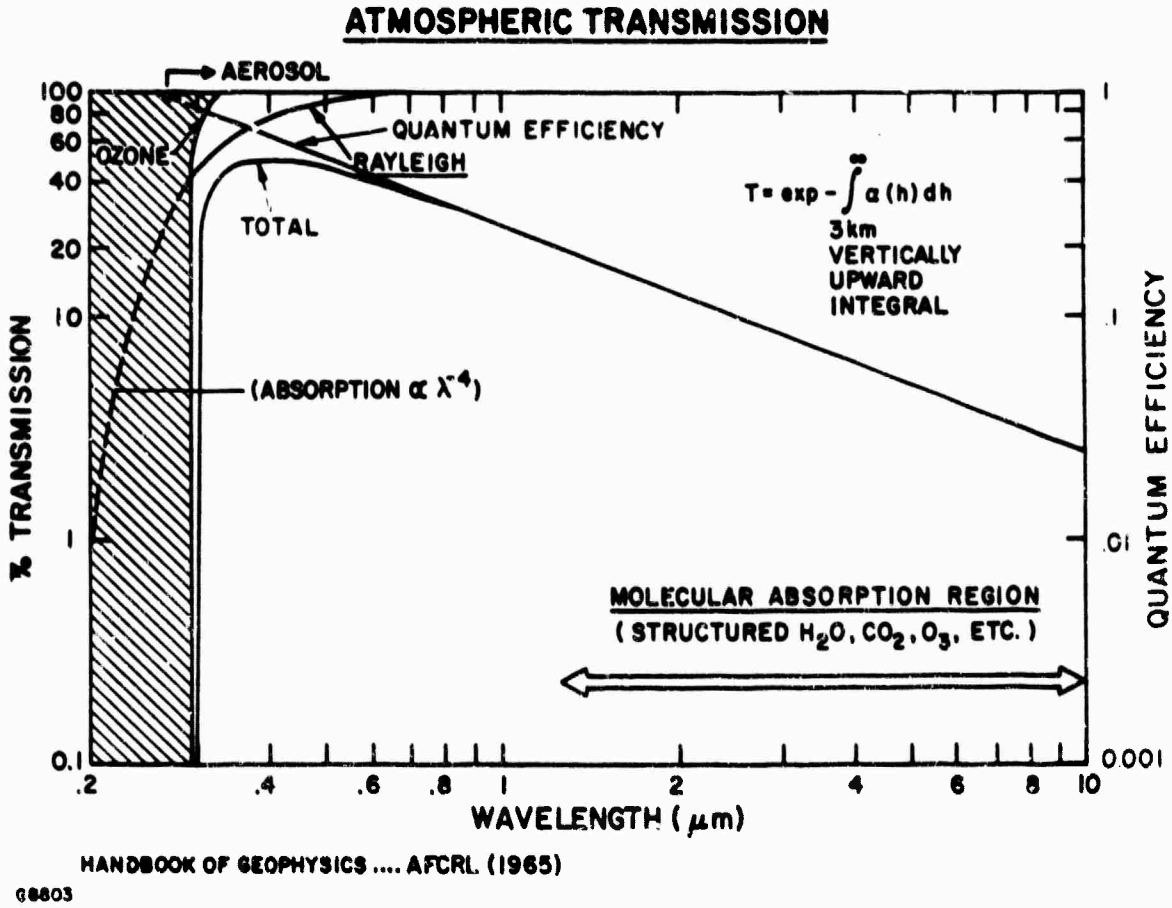

Figure 2 Propagation of Converted KrF Photons

been converted to a series of UV-visible lines due to vibrational Raman processes in high pressure molecular gases (e.g., H_2) showing good overall conversion efficiency[4]. In view of the above, it seemed reasonable and important to pursue scalable techniques that could efficiently convert KrF laser output to longer wavelengths. Two nonlinear optical conversion techniques that we had considered to achieve these goals were stimulated Raman and parametric conversion processes.

For the stimulated Raman process, phenomenologically, the acceptor atom can be thought of as absorbing an incident KrF photon thereby making a transition to an excited virtual state and then, with the emission of a Raman photon at longer wavelengths, proceeding to a level near the ground (initial) state. Through collisions with an efficient quenching gas, it can return to the initial state for subsequent re-excitation by the KrF laser field, i.e., exhibit high efficiency by recycling the metal atoms. The Raman process is enhanced when the virtual state is close to a real state.

Another method of "down conversion" to lower energy, longer wavelength photons applicable to UV laser light is parametric down conversion. In this process, conversion is achieved by the utilization of the non-linear properties of the medium (the acceptor atom or molecules). Here an atom in state 0 upon exposure to KrF laser light of frequency ν_1 goes to a virtual state 1 and re-emits three photons of frequencies ν_2, ν_3 and ν_4 such that $\nu_1 = \nu_2 + \nu_3 + \nu_4$. At the end of this process, the atom returns to its initial state entirely by optical transitions. Once again, if the various atomic transitions (ν_1, ν_2, ν_3 and ν_4) in the acceptor are allowed and the dipole moments are large, near resonant effects enhance the overall process such that efficient down-conversion should be likely.

At AERL under this contractual effort, theoretical and experimental research have been carried out on potentially efficient scalable schemes for converting KrF photons to longer wavelengths. By theoretical calculations, we have identified a number of promising candidates to convert the KrF laser radiation to longer wavelengths using stimulated Raman and parametric processes.

In that many of the candidates identified by this program as useful conversion systems are refractory metals (see Table 1), it was the approach of this contract to generate the needed densities by the dissociation of organometallic compounds e.g., $Pb(CH_3)_4$, cycla-pentadienyl trimethyl platinum, $Fe(CO)_5$, etc. The results of these experiments were quite successful and have been summarized in an earlier semi-annual technical report[5]. In addition, the theoretical effort provided needed calculations of the stimulated Raman emission cross sections for the various atomic acceptor candidates[5] and also the nonlinear susceptability for the parametric conversion processes.[6]

(4) Loree, T.R., Sze, R.C. and Barker, D.L., Appl. Phys. Letts., 31, 37 (1977).

(5) Trainor, D.W. and Mani, S.A., Optical Conversion Processes, Contract No. N00014-76-C-1162, Semi-Annual Technical Report, 15 Sept. 1976 to 15 March 1977.

(6) Trainor, D.W. and Mani, S.A., Optical Conversion Processes, Contract No. N00014-76-C-1162, Semi-Annual Technical Report, 15 March 1977 to 15 Sept. 1977.

TABLE 1. POTENTIAL CONVERSION CANDIDATES AND THEIR OUTPUT WAVELENGTHS

Candidate	Conversion Process	Output Wavelength λ, nm
Iron	Stimulated Raman	300, 304
Calcium	" "	544
Palladium	" "	332
Platinum	" "	322
Lead	" "	309
Hydrogen	" "	277, 313, 360
Thallium	Parametric Down Conversion	~ 380
Lead	" "	~ 364
Mercury	" "	~ 376
Lithium	" "	~ 680

Under this program, we modified a commercial KrF laser (Tachisto, Needham, MA) to provide a focused output of nearly 10^9 watts/cm^2 and used this laser to convert to wavelengths near 300 nm using atomic iron as the acceptor candidate[7]. A technical paper describing this work is included as Appendix A. In addition, we were able to demonstrate laser action in the organo-metallic precursor (i.e., the $Fe(CO)_5$). This represents a likely laser pumped photodissociation process followed by inversion of a photofragment produced in the initial step. A technical paper describing this system is included in this report as Appendix B.

We were in the process of evaluating other stimulated Raman candidates as well as beginning experimental investigations into lasing the parametric candidates when the funding for this technical program expired. The status of these experiments is summarized in the following sections of this final report.

(7) Trainor, D.W. and Mani, S.A., 30th Annual Gaseous Electronics Conference, Palo Alto, California, 20 Oct. 1977, Paper LA-3.

II. THEORETICAL CALCULATIONS

In addition to the calculations reported in the previous two semi-annual reports,[5, 6] further theoretical work was carried out on the parametric down conversion of KrF using lead atoms in the gaseous state as the acceptor candidate. The calculation of the non-linear susceptibility, $\chi^{(3)}$, proceeds in a fashion akin to that outlined in Ref. 6. The energy levels involved in the parametric process are shown in Figure 3. Although the relevant parametric process starts from the $^3P_1$ metastable state of the Pb atom, there will be a large population of the ground state $^3P_0$ atoms, which while not participating in the parametric process, contribute to the linear refractive index of the waves. This feature about lead allows phase matching to be achieved intrinsically for collinear propagation of the four waves. Figures 4, 5, and 6 give the phase mismatch as a function of output wave number for the $^3P_0$, $^3P_1$ and $^3P_2$ atoms respectively. Assuming Boltzmann equilibrium between the various levels at a given temperature, it is possible to calculate the output wave number as a function of temperature for exact phase matching. This is shown in Figure 7.

Calculations of the conversion efficiency for Pb were discussed in Ref. 6. Compared with Pb, both Li and Tl appear to be better candidates for ease of demonstration of lasing using the parametric down conversion of KrF laser radiation.

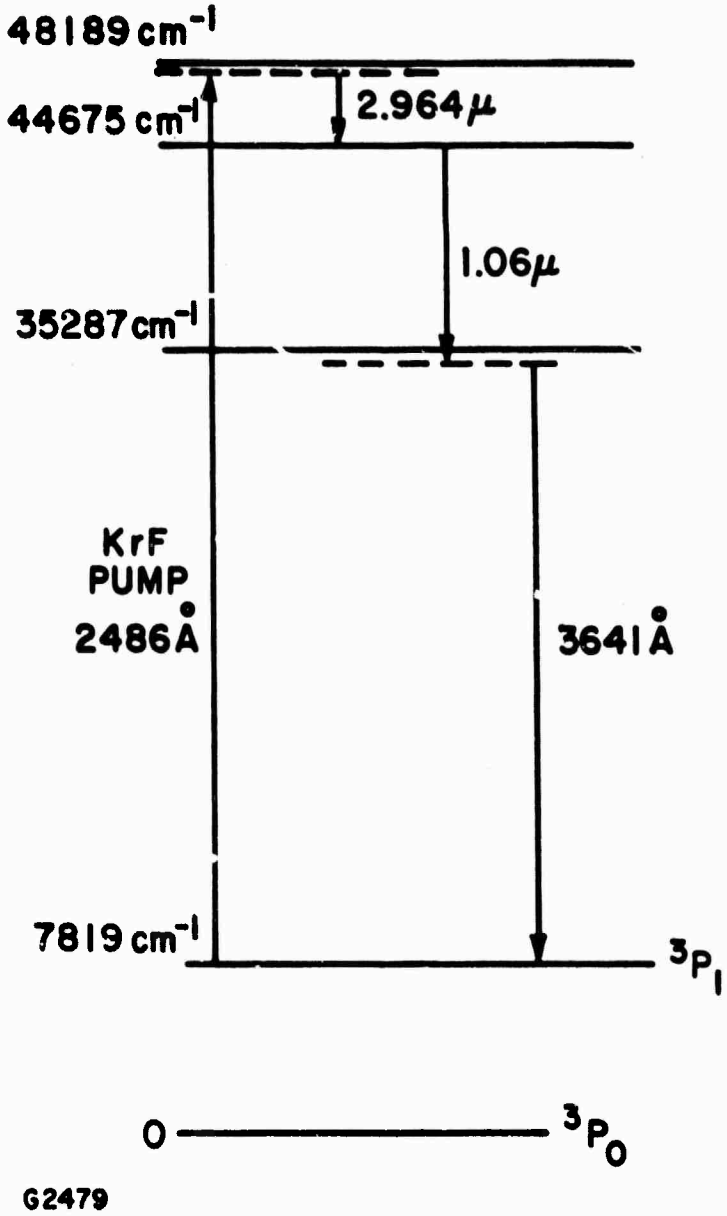

Figure 3 Energy Levels in Lead

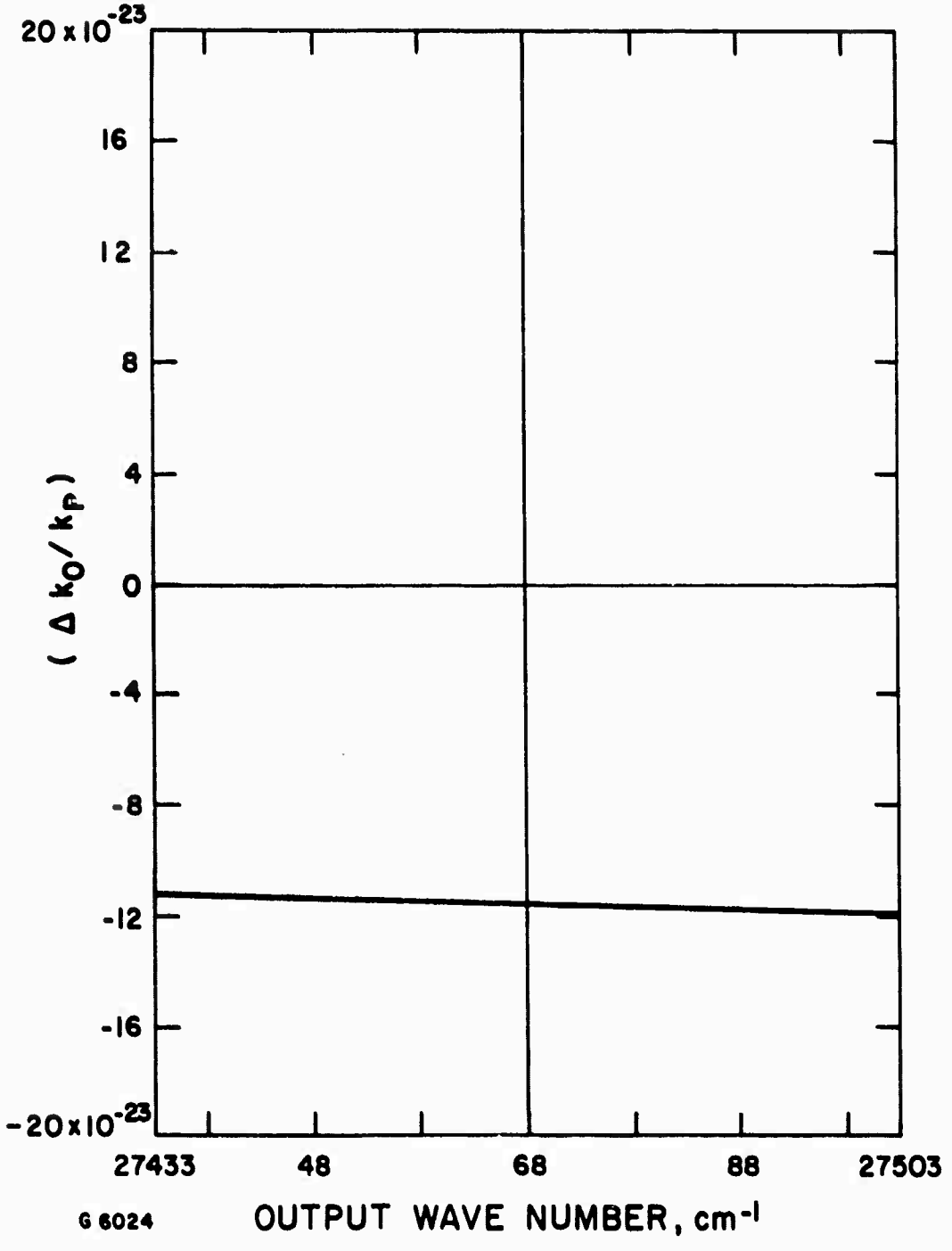

Figure 4 Phase Mismatch Δk (Normalized to Pump Wave Number k_p) vs Output Wave Number for $^3P_0$ Lead

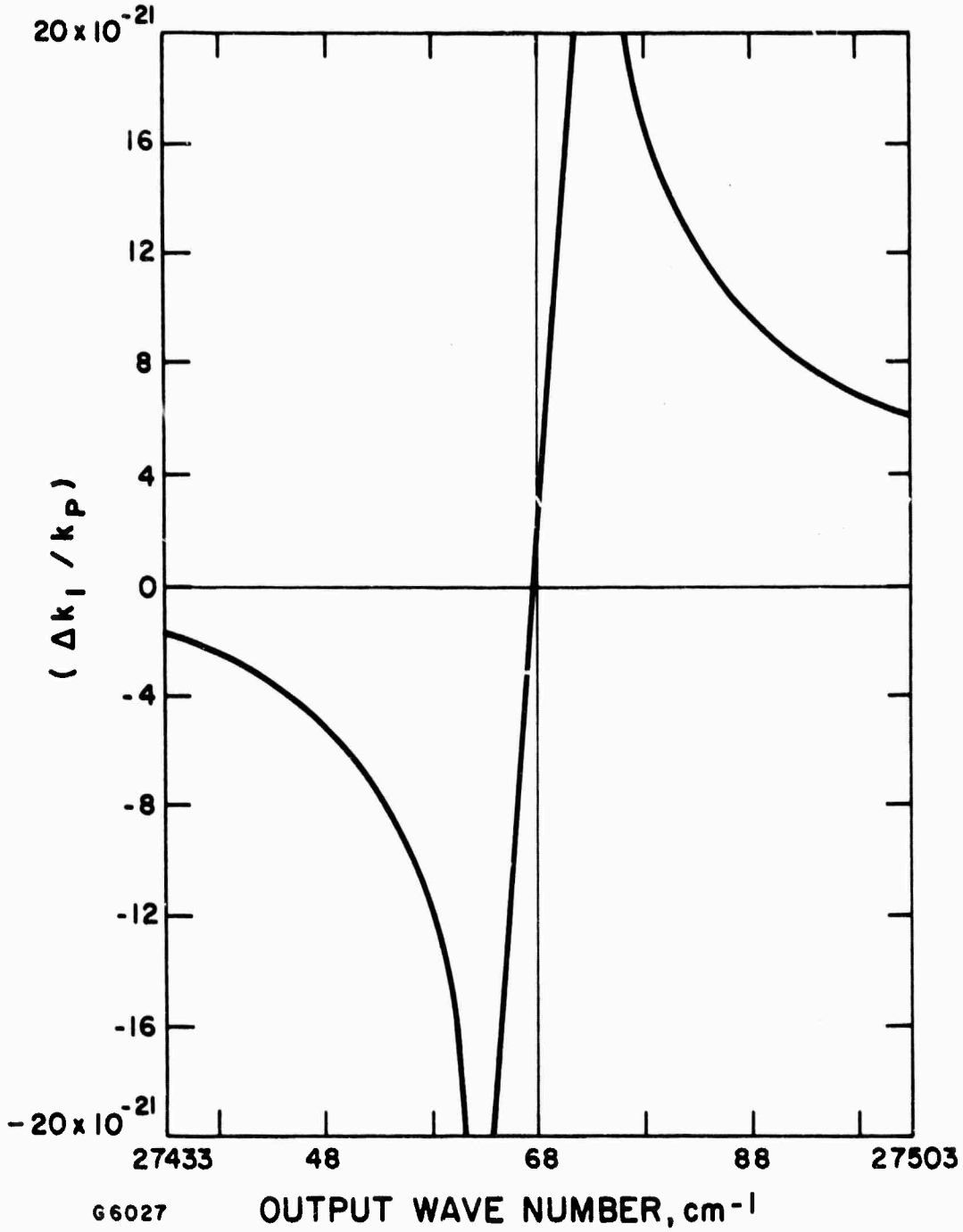

Figure 5 Phase Mismatch Δk (Normalized to Pump Wave Number k_p) vs Output Wave Number for $^3P_1$ Lead

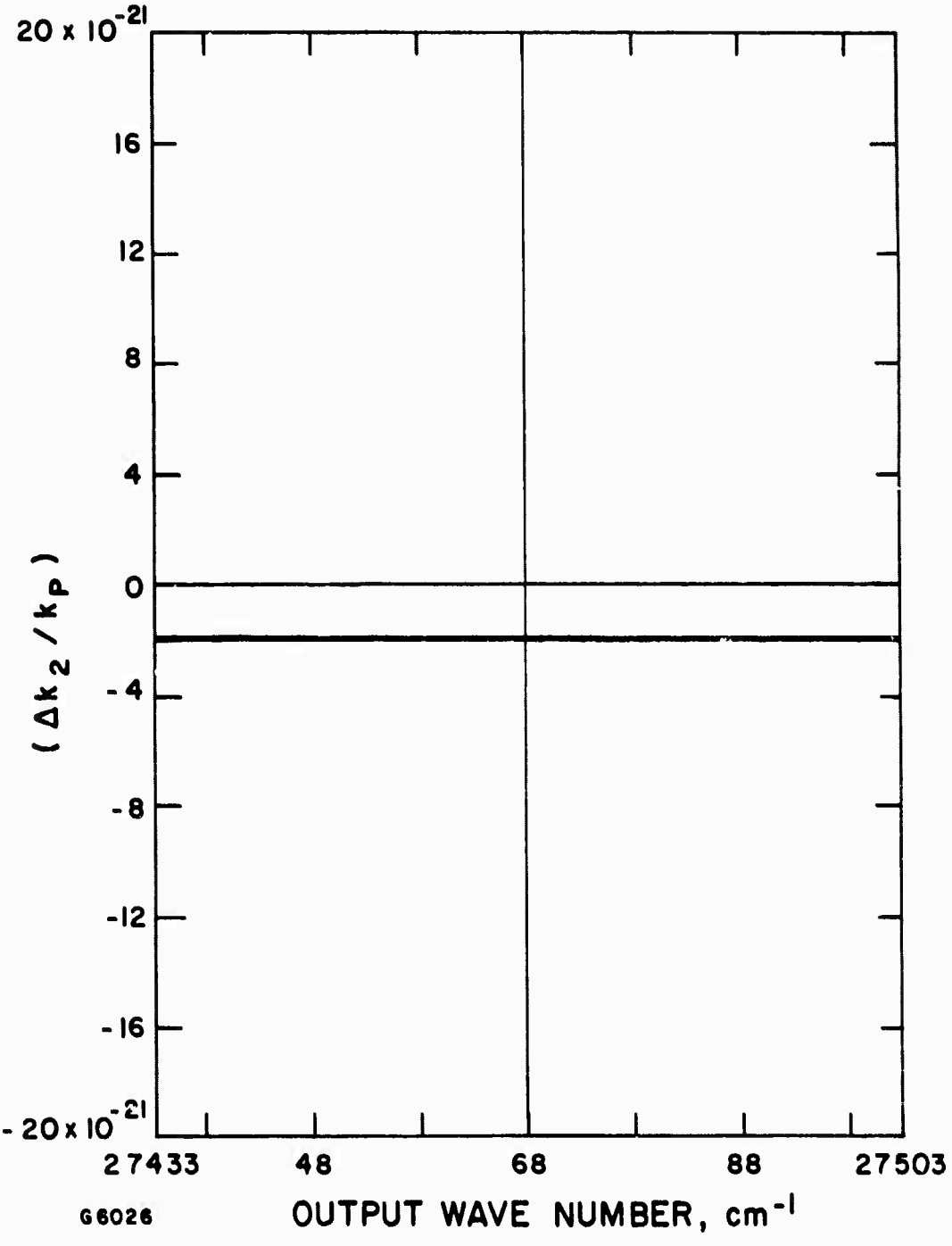

Figure 6 Phase Mismatch Δk (Normalized to Pump Wave Number k_p) vs Output Wave Number for $^3P_2$ Lead

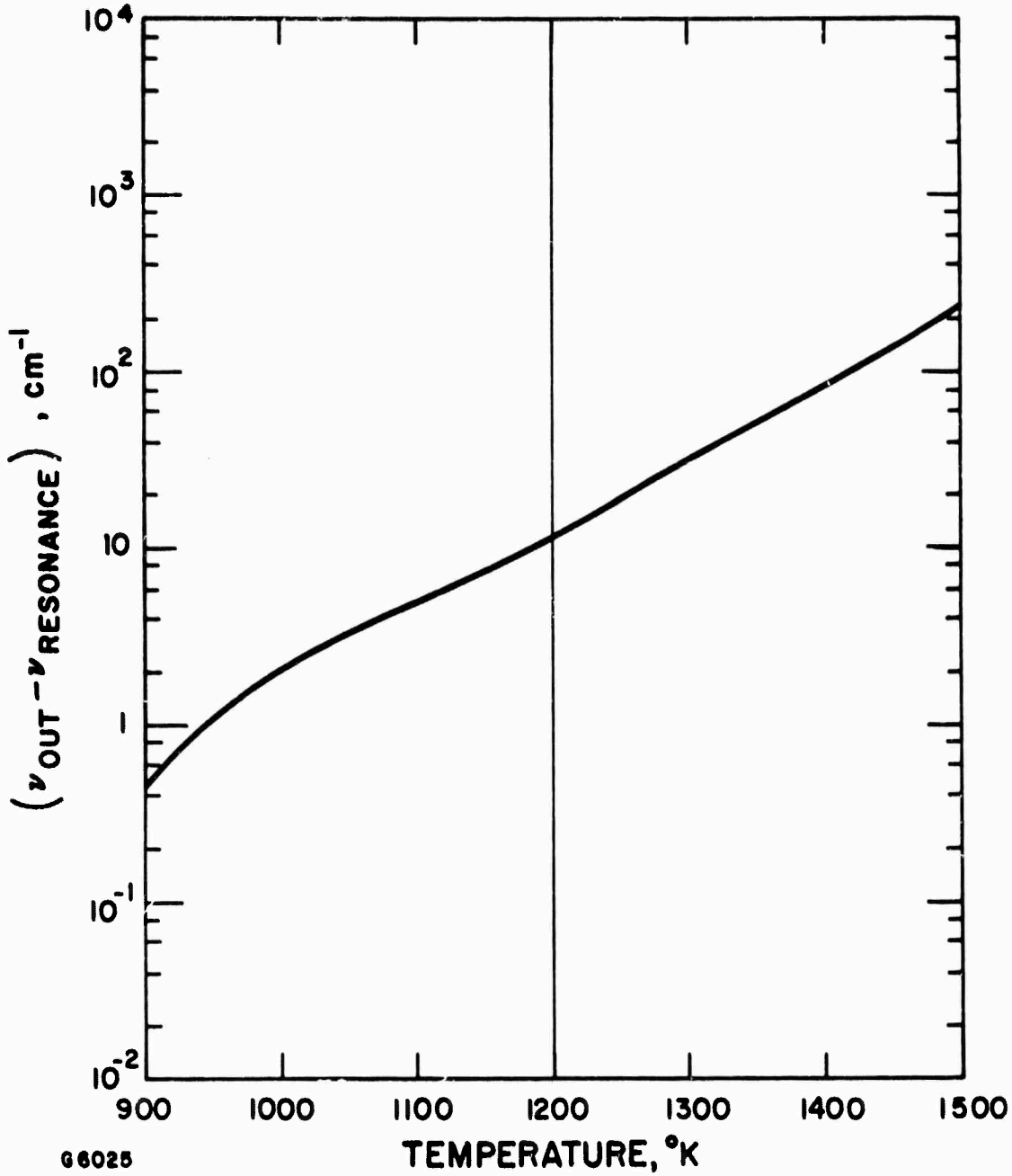

Figure 7 Detuning for Phase Matching vs Temperature

III. CANDIDATE EVALUATION AND STATUS

A. PLATINUM

Atomic platinum offered the potential of shifting the KrF output to propagating wavelengths with good quantum yield (see Figure 8). We have demonstrated the ability to produce reasonable densities of atomic platinum by the discharge dissociation of platinum acetyl acetonate in 50 torr of neon,[5] however, the disadvantages for demonstration of lasing proved to be difficult to overcome with the pump laser available. As discussed in our earlier semi-annual technical reports,[5, 6] a figure of merit to calculate total gain for a diffraction limited beam is the pump laser power divided by the bandwidth. For the modified Tachisto, its likely to be near 2×10^6 watts over ~ 100 cm^{-1}. For these laser performance characteristics and the achievable atom densities (> 5×10^{13} atoms/cm^3), the total stimulated Raman gain for the atomic platinum is calculated to be less than one. Demonstration of lasing would, therefore be likely only if improvements in the pump laser could be realized or a cavity made for enhancement of stimulated emission.

As in the case of iron,[5] platinum also has the potential for possible direct optical pumping, in addition to near-resonant stimulated Raman pumping. Since the bandwidth in the Tachisto pump laser extends over several Angstroms, some part of the laser output is in resonance with an atomic resonance line of platinum. We looked quickly, therefore, for some evidence of conversion via this mechanism. The results of these experiments showed no evidence of stimulated emission in the visible or near UV wavelength regions. Continued effort with platinum would require some improvement in the output power and/or bandwidth of the pump laser. For example, the performance characteristics of a 1 meter KrF laser developed at AERL[8] are 1.7×10^8 watts over 35 cm^{-1} and lasing demonstration using this device would be significantly more tractable for many of these candidates.

B. PALLADIUM

This conversion candidate is similar in many respects to platinum in that calculations suggest the possibility of stimulated Raman conversion of KrF to wavelengths near 332 nm. We have purchased an organo-metallic precursor to use for the production of atomic palladium, namely palladium (II) 2,4-pentanedionate (Alfa products). These experiments were about to be undertaken using the discharge production technique.

C. CALCIUM

This candidate offers the possibility of thermal production of reasonable densities and conversion to a visible wavelength (554 nm). There should

(8) Mangano, J.A., Jacob, J.H., and Hsia, J.C., 9th Winter Colloquium on Quantum Electronics, Park City, Utah, February 14-18, 1977.

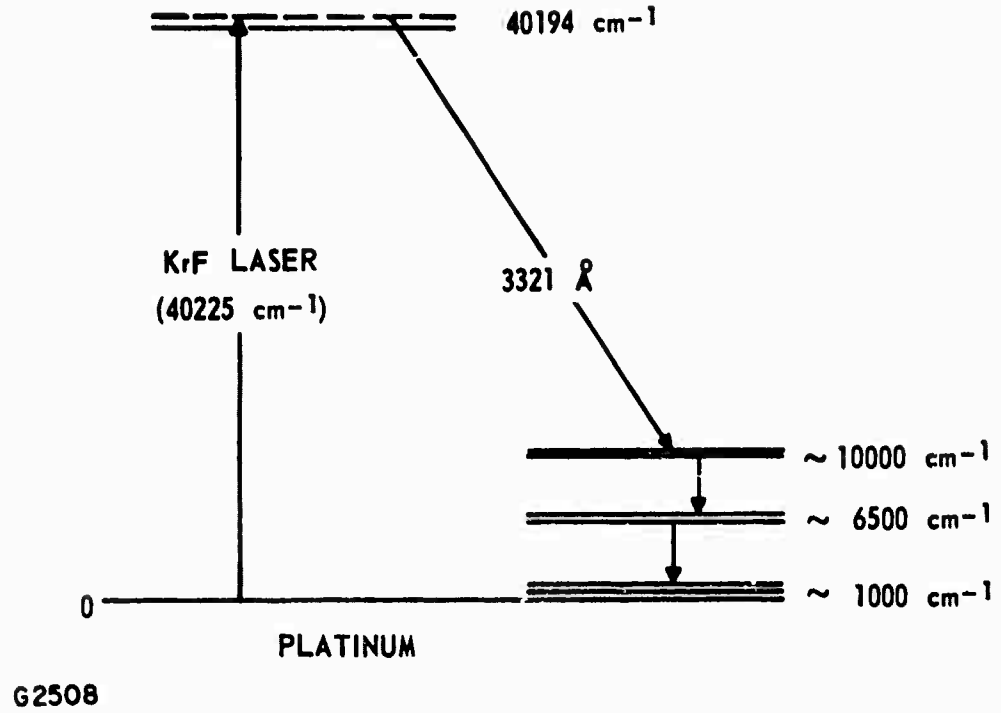

Figure 8 Schematic of KrF Pumped Platinum Stimulated Raman Laser

be no competing direct (resonant) pumping as KrF is approximately 1500 cm^{-1} off resonance from the scattering state participating in the Raman process. A test heat pipe was constructed from quartz using a stainless steel screen as a wick for the working fluid. Calcium pellets (Alfa products) were placed in the center of the heat pipe and this central section was heated to nearly 1300°K over a period of several hours. Since the Raman pump transition is far from any resonance, to achieve high total gains, substantial quantities of calcium is needed;[6] temperatures near 1500°K would thereby offer considerable improvement. This, however, is outisde the working range of a quartz heat pipe. A heat pipe constructed from steel would be necessary for additional study. This heat pipe was designed and was in the process of fabrication.

D. THALLIUM

This parametric candidate is also amenable to thermal production and, in anticipation of experimental production in a heat pipe, thallium metal was purchased for these investigations. A complete discussion of the relevant cross sections, susceptability, phase matching, etc. for thallium was presented in Ref. 6.

E. LITHIUM

Another attractive candidate for KrF conversion is atomic lithium, where once again experimental investigations can be conducted producing lithium by thermal sources. Lithium metal was purchased and a heat pipe was designed for use in these experiments.

F. IRON PENTACARBONYL

This lasing compound was discussed in some detail in a previous semi-annual report[6] and a technical paper is included as Appendix B of this final report. This compound offers the possibility of efficient laser output at a variety of wavelengths. It also represents an example of a class of compounds which could show similar characteristics (e.g., $Ni(CO)_4$, $Cr(CO)_6$, etc.). Experiments to measure conversion efficiency in a transverse pumped geometry were being undertaken as forward scattered measurements were not an ideal pumping configuration. Additional investigation into the effect of varying the ligands of the parent compound on the output wavelength may also prove rewarding. The results of these experiments suggest the possibility of greatly simplifying the overall project with regard to the engineering aspects of a large scale converter. More work is needed in this new area.

IV. SUMMARY

Considerable success in the identification, evaluation, production and demonstration of lasing in a number of optical conversion candidates has been attained. Viable candidates to successfully convert the output of a high power KrF laser into more propagating wavelength regions have been identified and methods to scale to high average power have been considered. Lasing action in atomic iron and in iron pentacarbonyl has been demonstrated.

APPENDIX A

KrF LASER PUMPED ATOMIC IRON LASER

APPENDIX A

KrF LASER PUMPED ATOMIC IRON LASER*

Daniel W. Trainor and Siva A. Mani

ABSTRACT

A technique for producing stimulated emission in an optically pumped atomic iron system at room temperature is described. The required iron density (~ 10^{14} atoms/cm^3) for single pass amplified spontaneous emission was produced at room temperature by two techniques: a low pressure (50 torr) discharge of iron pentacarbonyl and neon and by the flash photo-decomposition of $Fe(CO)_5$ in an argon buffer. A commercial KrF laser producing output powers of up to 1 MW was modified to improve the beam quality, and the beam was focused into the reaction cell. Resonant processes involving the $3d^6$ 4s 4p ($^5F^o$) intermediate state of iron and the KrF laser field ($\lambda \approx 248$ nm) produced stimulated emission near 300 and 304 nm in agreement with theoretical predictions.

*A preliminary report of this work was given at the 30th Annual Gaseous Electronic Conference, Palo Alto, California, 20 October 1977, Paper #LA-3.

I. INTRODUCTION

Over the past few years, a number of new lasers have been reported, lasers that are similar in their formation kinetics and the nature of the states involved in the lasing transition. These include the rare gas monohalides,[1-6] molecular halogen,[7-9] and mercury halide[10, 11] lasers. Many of these laser systems possess strongly bound, ionic upper states and weakly bound or even repulsive, covalent lower states. Typical of each of these classes are ArF (193 nm),[12] I_2 (342 nm)[8] and HgCℓ (558 nm).[10] Since their initial lasing, many of the ultraviolet lasers have been shown to be efficient, scalable, high power laser systems and therefore considered for a variety of practical applications. Many such applications require alternate wavelengths, and experimental and theoretical efforts were undertaken to shift their outputs into more useful regimes.

In order to convert these lasers output to longer wavelengths, some acceptor candidate is necessary to provide a perturbation so as to alter the scattered photons energy. For stimulated Raman concepts, molecules may provide vibrational states for conversion whereby the scattered photon loses (Stokes) or gains (anti-Stokes) energy from the vibrational contribution. These processes occur through a virtual state that is far from any resonant state. In atomic electronic Raman scattering or direct optical pumping, increased cross sections can be realized if the virtual state is near-resonant or resonant with a real state and is accessible via allowed transitions from

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(6) Ault, E.R., Bradford, R.S. and Bhaumik, M.L., Appl. Phys. Lett. 27, 413 (1975).

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(10) Parks, J.H., Appl. Phys. Lett. 31, 192 (1977).

(11) Parks, J.H., Appl. Phys. Lett. 31, 297 (1977).

(12) Hoffman, J.M., Hays, A.K. and Tisone, G.C., Appl. Phys. Lett. 28, 538 (1976).

a populated initial state (e.g., the ground state). Another consideration, besides seeking a near-resonant transition at the pump laser wavelength, is the energy of any scattered photon, i.e., does the real state couple to any lower lying states via transitions with high probability and at desirable wavelengths.

For example, Djeu and Burnham[13] converted the output of an XeF laser (351 nm) to near 585 nm using electronic stimulated Raman techniques with barium vapor produced in a heat pipe. Loree, Sze, and Barker[14] have used molecular Raman transitions to shift the output of ArF (193 nm) and KrF (248 nm) to a series of lines in the region 190-360 nm using high pressure gases (e.g., H_2). In addition to the Raman experiments, Burnham[15] has reported stimulated emission from atomic indium at 451 nm by the photodissociation of indium monoiodide vapor using an ArF laser. Recently, Schimitschek, Celto and Trias[16] have observed molecular electronic inversion on the $B^2\Sigma^+ \rightarrow X^2\Sigma^+$ transition of the HgBr radical by photodissociating $HgBr_2$ with an ArF laser. In our laboratory, we have observed lasing in iron pentacarbonyl when irradiated by a high power KrF laser.[17] For KrF conversion (248 nm), many atomic acceptor candidates are refractory metals, e.g. iron, platinum, etc.

To produce sufficient densities of refractory metals for conversion experiments, we opted to utilize techniques involving the decomposition of organo-metallic compounds. Earlier experimentation showed that reasonable densities of a number of metals could be produced by the flash photodecomposition of compounds like $Pb(CH_3)_4$,[18,19] $Bi(CH_3)_3$[20,21] and $CuC\ell$ or CuBr.[22] Iron pentacarbonyl had been similarly utilized to provide kinetic rate constant information on the J-state sublevels of the $^5D^o$ states of atomic iron.[23]

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(13) Djeu, N., and Burnham, R., Appl. Phys. Lett. 30, 473, (1977).

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(15) Burnham, R., Appl. Phys. Lett. 30, 132 (1977).

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We report here the results of these experiments whereby single pass amplified spontaneous emission from atomic iron produced from the decomposition of $Fe(CO)_5$ and pumped by a KrF laser was observed. Resonant processes involving the $3d^6\ 4s\ 4p\ (^5F^o)$ intermediate state of iron and the KrF laser field (248 nm) produced stimulated emission near 300 and 304 nm in agreement with theoretical predictions.

II. EXPERIMENTAL

Our flash photolysis equipment has been described in detail in earlier publications[18-22] (see Figure A-1). It consists of a 45 cm long reaction cell (1.8 cm I.D.) constructed of suprasil quartz with quartz windows fused to the cell itself. Surrounding and parallel to the reaction cell are 8 linear suprasil quartz flashlamps. Typical energy ($1/2\ CV^2$) supplied to each lamp is 50 joules.

In addition to producing atomic iron using flash photolysis techniques, we used low pressure glow discharges of $Fe(CO)_5$ in neon to produce the needed iron densities. The discharge cell was constructed by fusing together two metal to glass seals and attaching window holders (see Figure A-2). In this way, the metal ends acted as electrodes for the discharge which was produced by dissipating the energy stored in a 0.02 μf capacitor charged to 20 kV. This approach of using discharge dissociation of organo-metallics has been used by Chou and Cool[24] to produce new laser transitions at 24 wavelengths from 9 different metals. Also, Gabai et al.[25] used discharge dissociation techniques to homogeneously oxidize acetylene or cyanogen containing tetramethyl lead to produce significant populations of lead metastable states. It is a versatile technique applicable to many similar systems.

The pump laser used in these experiments was a discharge initiated KrF excimer laser (Tachisto Corp., Needham, MA). As delivered, the laser provided approximately 50-60 mJ of 248 nm photons in a 20 nsec pulse with a stated beam divergence near 4 mrad. One and two shot exposures of the output were analyzed by microdensitometer traces of a photographic plate to obtain the spectral width and it was found to be near 10Å ($\sim 160\ cm^{-1}$ fwhm). The output is rectangular approximately 15 mm x 4 mm. To achieve high flux, it was necessary to improve the laser beam quality so it could be focused tightly. This was accomplished using the techniques described by Barker and Loree[26] and consisted of replacing the supplied output coupling mirror with a 50 cm fl plano-convex lens (suprasil). This provided an unstable cavity configuration of good beam quality which focused nearly 50 cm from the Tachisto exit port. Typical output was near 30-40 mJ in 20 nsec which when focused provided nearly a gigawatt/cm^2.

A typical experimental arrangement is shown in Figure A-3 where the output of the KrF laser is shown focused into the discharge cell. The KrF output was monitored for shot to shot variations by a photodiode and the

(24) Chou, M.S. and Cool, T.A., J. Appl. Phys. 47, 1055 (1976).

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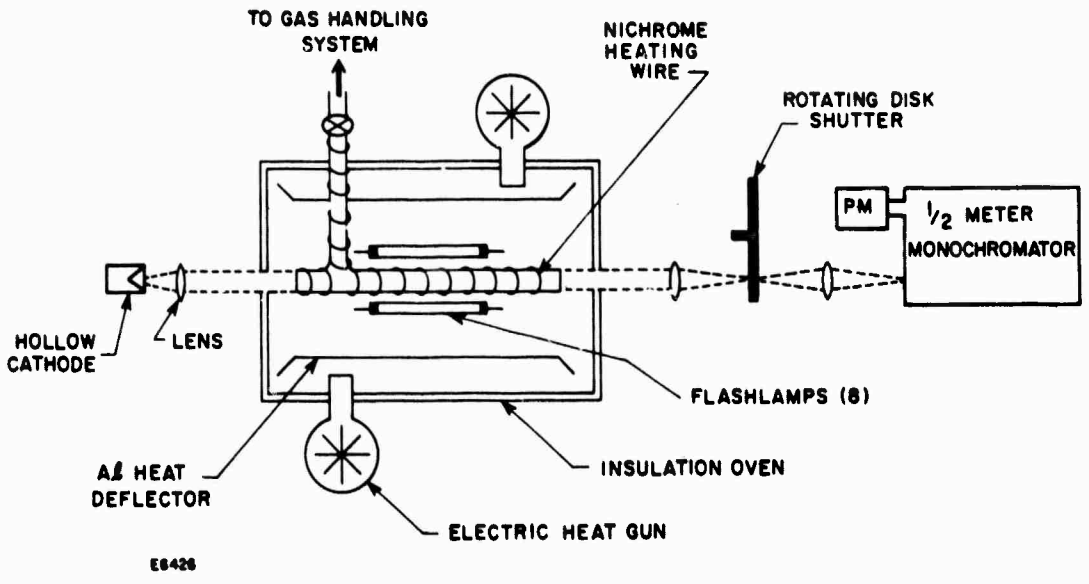
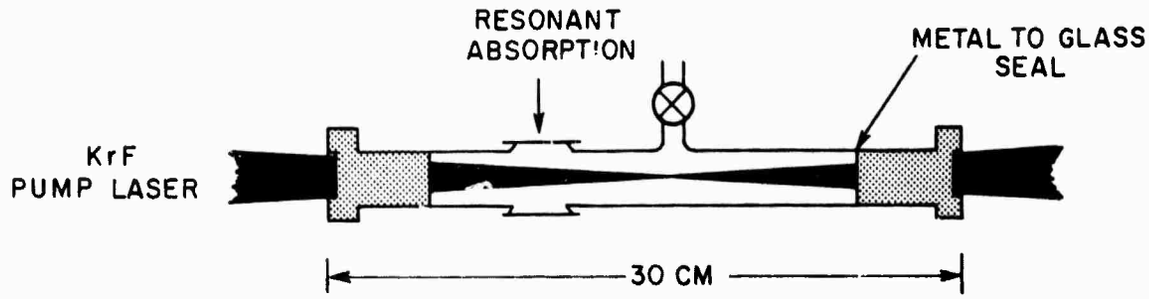
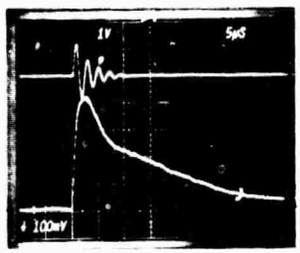


Figure A-1 Diagram of Flash Photolysis Apparatus



50 TORR NEON + 0.1 TORR ORGANO-METALLIC



CURRENT PULSE

BROAD BAND LIGHT EMISSION

H1933

Figure A-2 Discharge Cell Characteristics

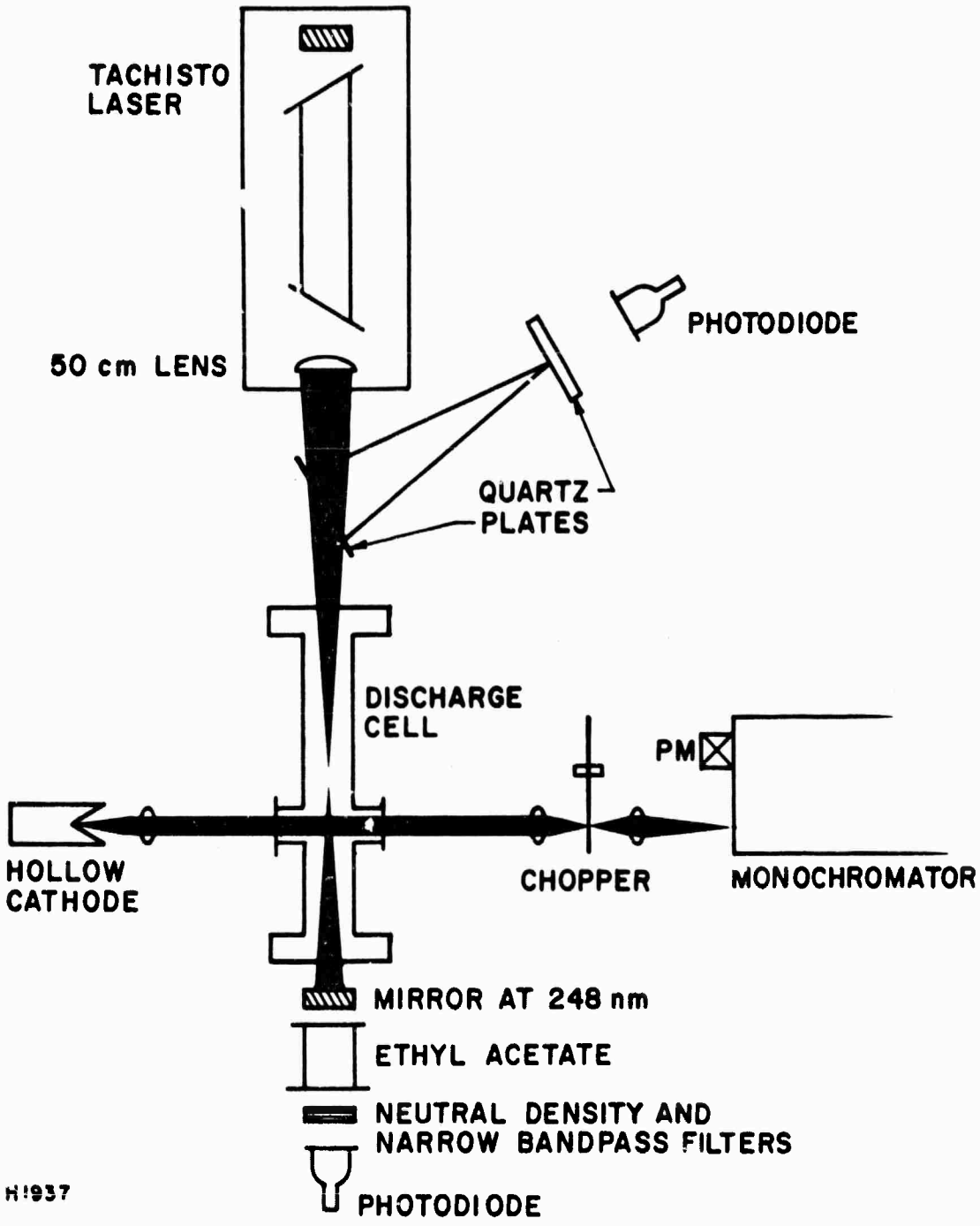


Figure A-3 Experimental Approach

total energy by an energy meter (Scientech model 362). The converted KrF photons were monitored in the forward scattered direction by a calibrated photodiode with discrimination against 248 nm being accomplished through use of a 99% reflecting mirror (248 nm), as well as chemical and narrow bandpass filters. Total signals below photodiode saturation levels were maintained through use of neutral density filters.

With the discharge or the flash reaction cell, the production and decay of any atomic iron produced could be monitored by using resonant absorption techniques. The light source consists of an iron hollow cathode (Glomax, Barnes Engineering) and a mechanical shutter, monochromater/photomultiplier detection system for monitoring the resonance line chosen for the time-resolved absorption measurements.[27] Typical ground state metal atom densities achievable were near 10^{13} - 10^{14} atom/cm^3 for the discharge and 10^{11} - 10^{12} atoms/cm^3 for the flash photolysis approaches respectively.

The iron pentacarbonyl (Alpha Products) was subjected to several freeze-pump-thaw purification cycles before dilution with the appropriate buffer gas. Neon (Cryogenic Rare Gas Laboratories, 99.99%) and argon (Liquid Carbonic, 99.98%) were used without further purification.

(27) Yatsiv, S. and Ewing, J.J., Rev. Sci. Instrum. 45, 705 (1974)

III. RESULTS AND DISCUSSION

As a check on our overall system, we opted to repeat the molecular Raman conversion experiments of the LASL group[14] using a 60 cm x 3.8 cm dia stainless steel cylinder filled to near 10 atm with hydrogen. We were able to observe 4 or 5 Stokes shifted lines as well as one anti-Stokes transition using a simple prism spectrograph and Polaroid film for observation.

Similar experiments were then undertaken with the atomic iron as the converter. Some typical data are shown in Figure A-4. From the atomic absorption data, it can be seen that the ground state ($^5D_4$) reaches a maximum density near 300 μsec following initiation of the discharge. By using suitable delay components, we fired the KrF pump laser at the time where the [Fe] was maximum. The pump laser time history is shown in the top trace and represents typically 30 mj of KrF photons. The converted output near 300 nm is shown in the same oscillogram and indicates a pulse of somewhat shorter duration. These and other data taken with the calibrated photodiodes showed that approximately 2% of the pump laser energy was converted to output near 300 nm. Additional converted output would require increased atomic iron density or the use of each atom produced more than once.

As a check on the optimum time to delay the laser relative to the discharge production, we probed the reaction cell with the laser pulse at varying times from the discharge. The results are presented in Figure A-5 where maximum converted output from the photodiode signal is seen to occur near 200 to 300 μsec in good agreement with the atomic absorption data.

Replacing the photodiode detector with a prism spectrograph showed the lasing to occur over four lines (see Figure A-6). This corresponds to atomic iron transitions[28] as shown in Figure A-7. We were also able to check the varying spectral output by probing the discharge at different delay times and then determining the intensities of the various lines through densitometer traces of the photographic plate. The data are plotted as intensity peak height (in arbitrary units) vs time on a semi-logarithmic scale in Figure A-8. From these data, it is readily seen that the 300 nm transition shows the greatest output and persists with decreasing iron density for longer times. This is consistent with the total gain calculated for this transition and reflects the higher density in the lowest spin-orbit state as well as its larger transition probability.

In addition to these experiments using the discharge cell, we investigated lasing when the iron was produced by the flash photodecomposition of iron pentacarbonyl. A comparison of flash vs discharge experimental results from typical experiments are shown in Figure A-9. Here the atomic

(28) Corliss, C.H. and Bozman, W.R., Nat. Bur. Stand. (US) Monograph No. 53 (1962)

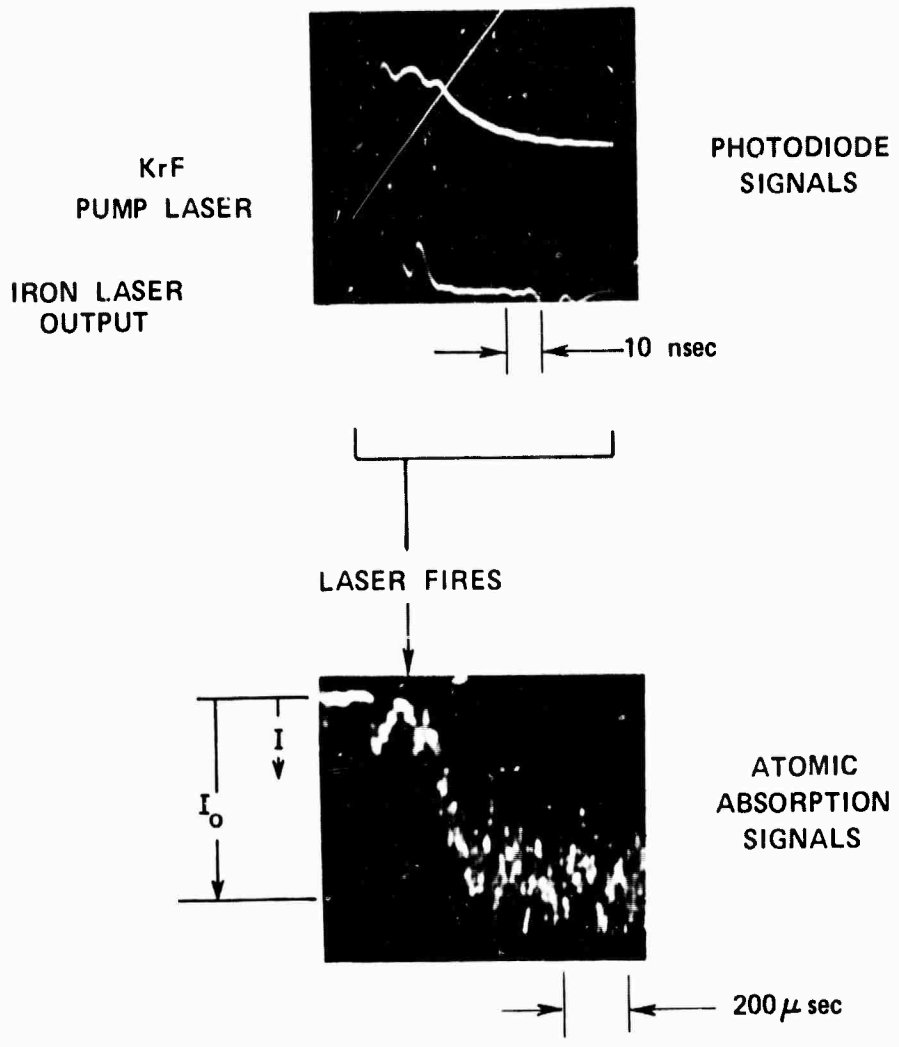


Figure A-4 Experimental Data

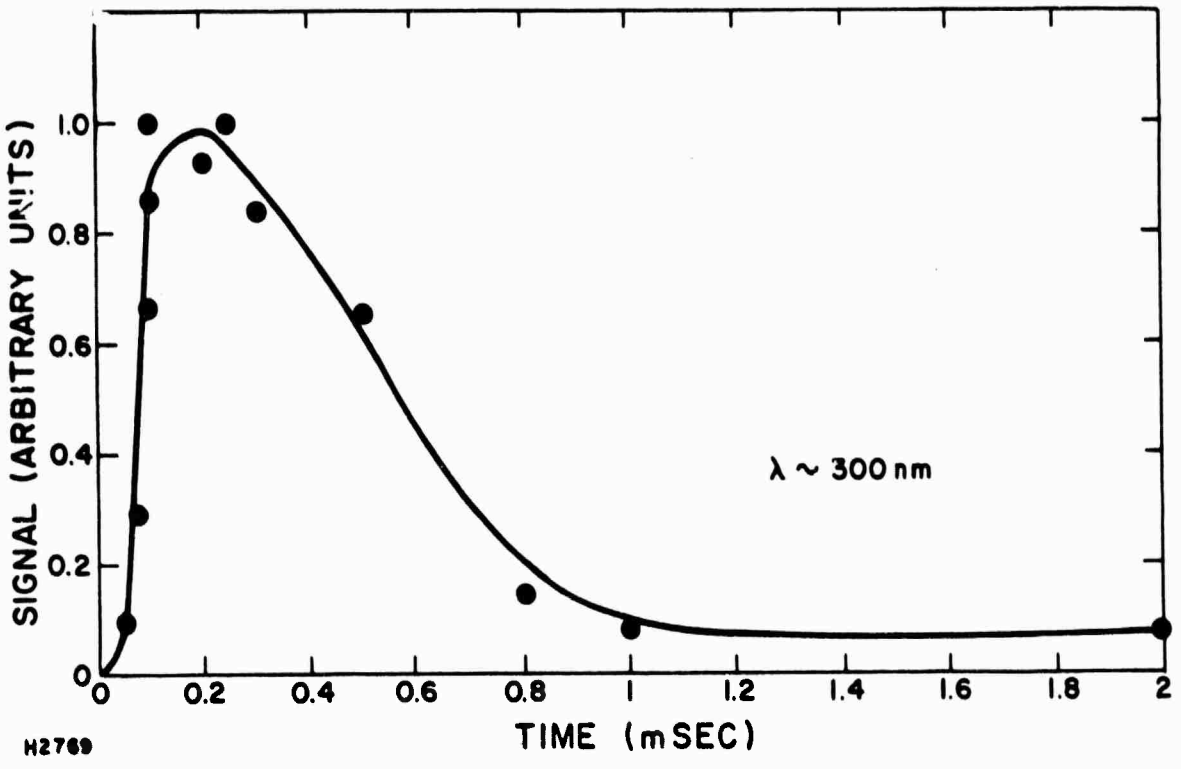


Figure A-5 Converted Laser Output vs Atomic Iron Density

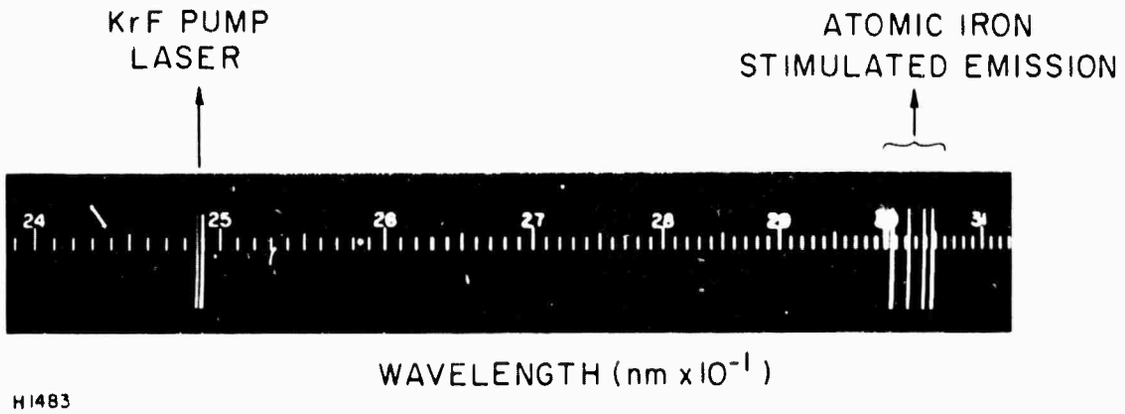


Figure A-6 Optically Pumped Atomic Iron Laser

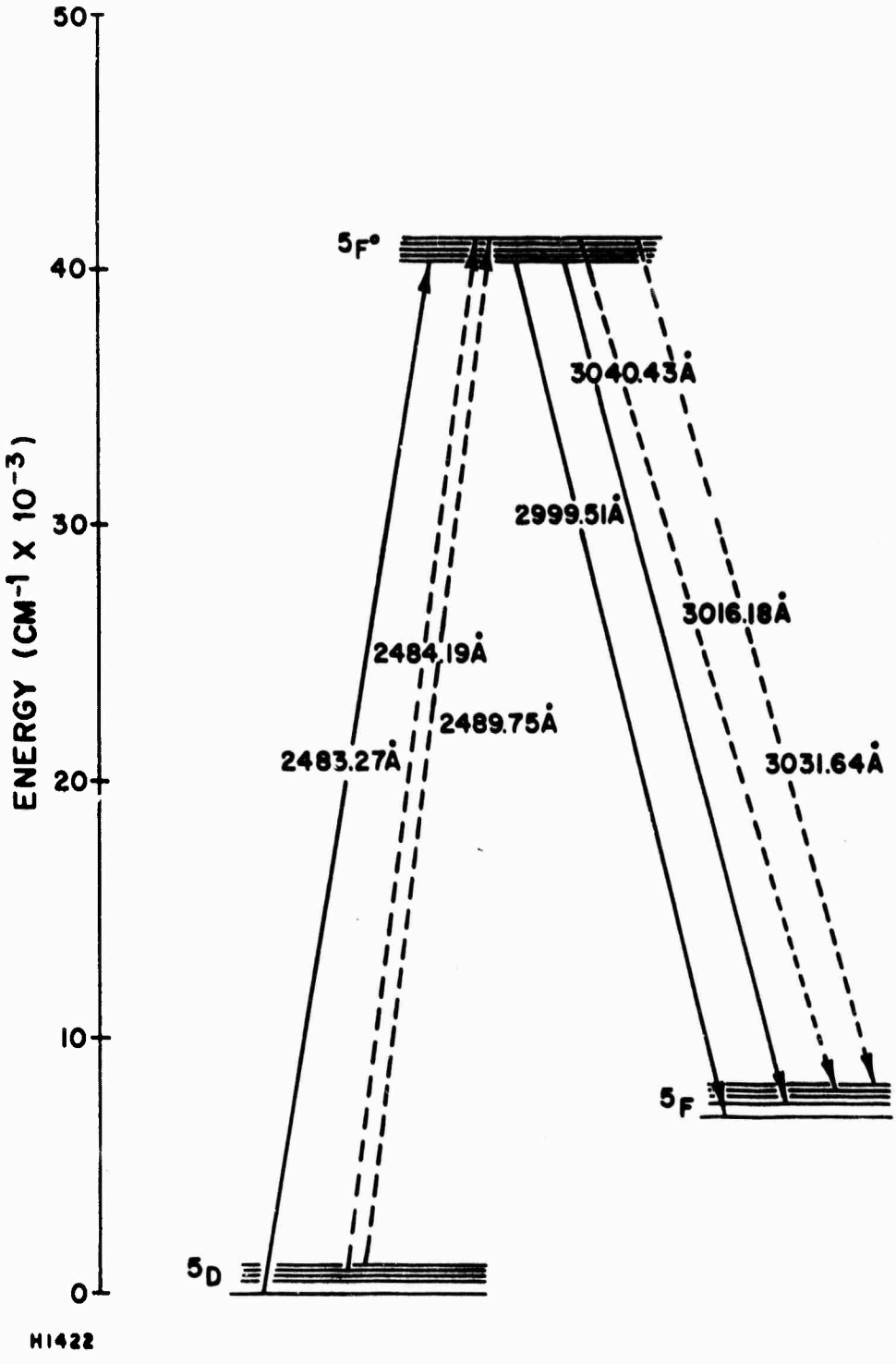


Figure A-7 Atomic Iron Lasing Transitions

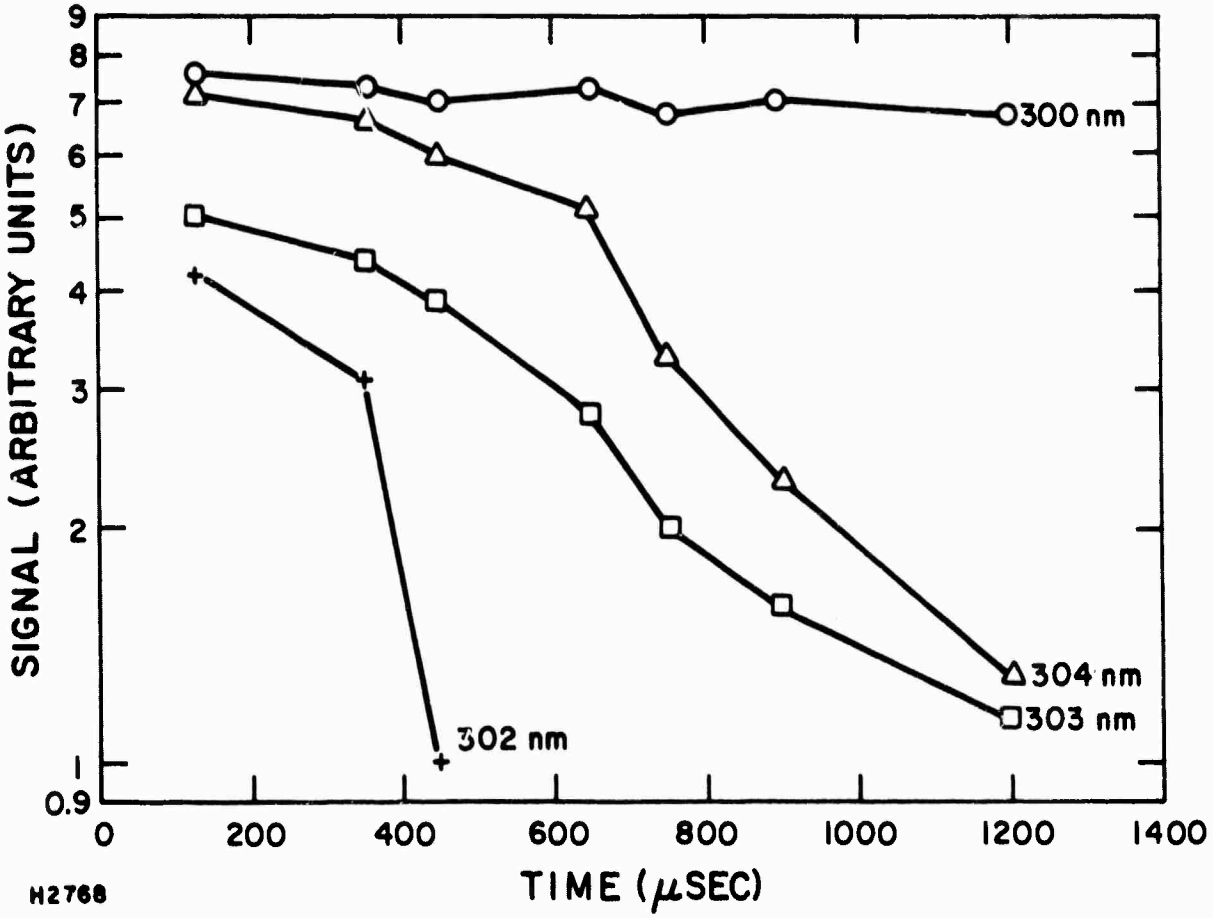


Figure A-8 Spectral Distribution as a f (Atomic Iron Density)

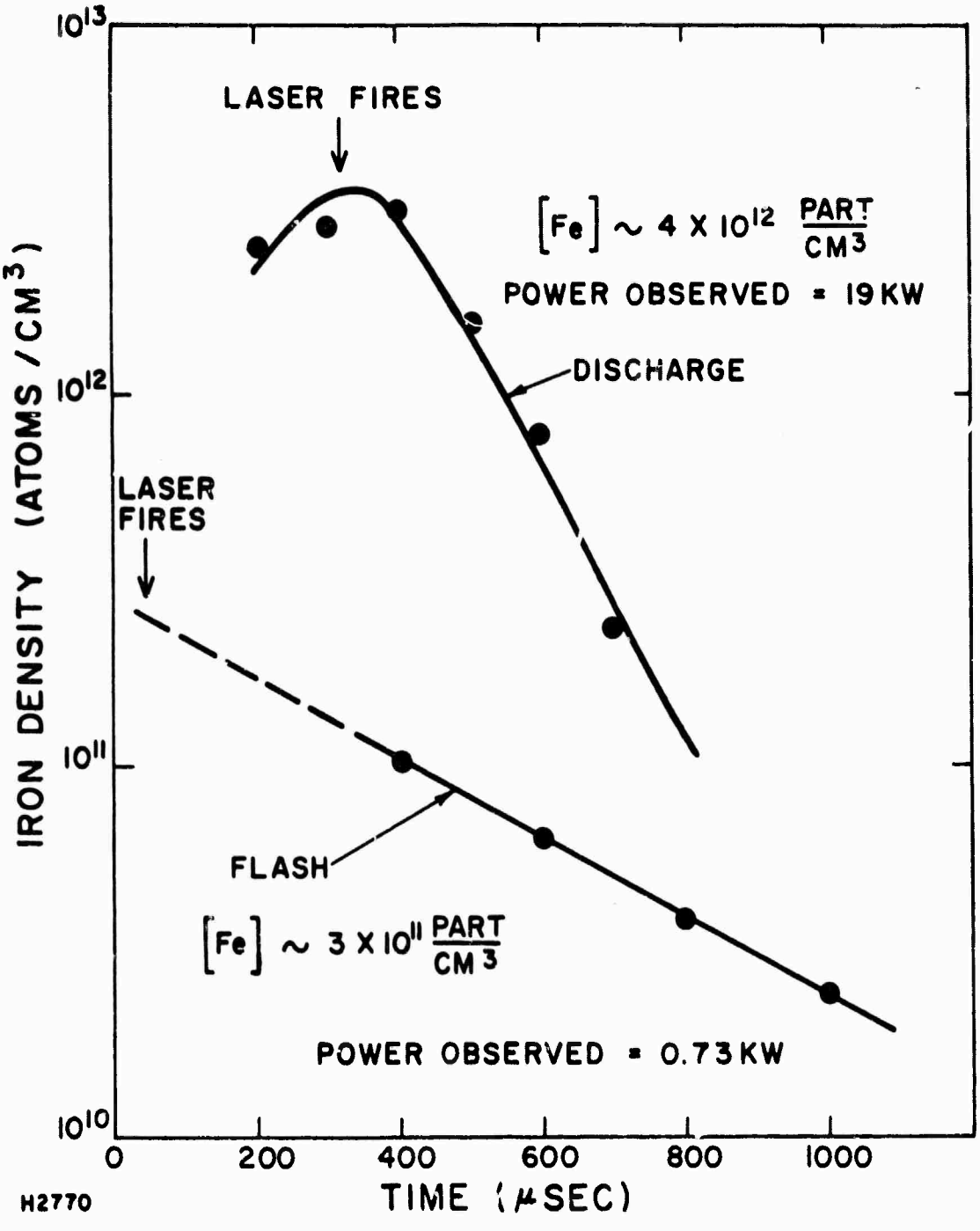


Figure A-9 Flash vs Discharge: Iron Laser Output

iron production is lower and the output is down correspondingly. Also, since the optimum $Fe(CO)_5$ density in these flash experiments was near 6×10^{12} cm^{-3}, we were able to show that the forward and backward scattered atomic iron lasing photons were comparable. In the discharge experiments, we operated with nearly 1×10^{16} cm^{-3} $Fe(CO)_5$ and at these densities absorption[29] of the pump laser light by the $Fe(CO)_5$ was considerable and therefore the solution to the gain equation for forward and backward scattering shows an asymmetry for these conditions, the forward gain being larger than the backward gain. This was verified experimentally.

(29) Lundquist, R.T. and Cris, M.J., J. Org. Chem. 27, 1167 (1962).

IV. SUMMARY

We have demonstrated successful wavelength conversion of KrF laser radiation by optical pumping in a refractory metal vapor at room temperatures. The metal vapor has been produced in a scalable manner by both flash photo decomposition and discharge decomposition of $Fe(CO)_5$. Single pass amplified spontaneous emission at 300 and 304 nm has been observed in agreement with theoretical predictions. This method of production of metal atoms can be applied to other refractory metals as well, and this approach opens up the possibility of development of a new class of optically pumped or Raman pumped lasers not conveniently accessable using thermal production techniques.

The authors gratefully acknowledge fruitful discussions with I. Itzkan, M. Rokni, J. Mangano and M. Kovacs during the execution of the experiment and the participation of A. Montagna and K. Wildnauer in collecting the experimental data.

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APPENDIX B

IRON PENTACARBONYL PHOTODISSOCIATION LASER

APPENDIX B

IRON PENTACARBONYL PHOTODISSOCIATION LASER

Daniel W. Trainor and Siva A. Mani

ABSTRACT

Laser action was observed when mixtures of $Fe(CO)_5$ and an inert gas were irradiated with a focused beam of KrF laser radiation (λ = 248 nm). Single pass amplified spontaneous emission was observed in two wavelength regions: the near UV and the yellow-green. Measurements made with narrow bandpass filters showed nearly 60% of the output radiation was at 385 nm.

Over the past year, a number of experimental results have been reported whereby high power excimer lasers have been used to produce laser action at other wavelengths. Djeu and Burnham[1] have converted the output of an XeF laser (351 nm) to near 585 nm using electronic stimulated Raman techniques with barium vapor produced in a heat pipe. Loree, Sze, and Barker[2] have used molecular Raman transitions to shift the output of ArF (193 nm) and KrF (248 nm) to a series of lines in the region 190-360 nm using high pressure gases (e.g., H_2). In addition to the Raman experiments, Burnham[3] has reported stimulated emission from atomic indium at 451 nm by the photodissociation of indium monoiodide vapor using an ArF laser. Recently, Schimitschek, Celto and Trias[4] have observed molecular electronic inversion on the $B^2\Sigma^+ \rightarrow X^2\Sigma^+$ transition of the HgBr radical by photodissociating $HgBr_2$ with an ArF laser. In our laboratory, we have observed lasing in atomic iron (300 nm) through optical pumping by a KrF laser.[5] The atomic iron was produced at room temperature by the dissociation of iron pentacarbonyl. During the course of this work, we observed single pass amplified spontaneous emission from the precursor used to produce the iron atoms, namely the $Fe(CO)_5$.

At room temperature, iron pentacarbonyl is a yellow liquid with a vapor pressure near 30 torr. In sunlight or under ultraviolet irradiation, it evolves carbon monoxide and dark yellow platelets of iron enneacarbonyl, $Fe_2(CO)_9$. The following mechanism has been proposed to explain these observations.[6]

$$Fe(CO)_5 + h\nu \rightarrow Fe(CO)_5^*$$

$$Fe(CO)_5^* + Fe(CO)_5 \rightarrow Fe_2(CO)_9 + CO$$

The infrared, Raman[7] and ultraviolet[8] spectra have been studied by a number of workers. Near KrF wavelengths (248 nm), it has an absorption cross section of 1.7×10^{-17} cm^2. It has been flash photolyzed to study spin-orbit relaxation among the J sublevels of the lowest energy state of atomic iron[9] and has been investigated in a novel approach to produce lasing in atomic iron by direct electron dissociative excitation.[10]

(1) Djeu, N., and Burnham, R., App. Phys. Letters 30, 473 (1977).

(2) Loree, T.R., Sze, R.C., and Barker, D.L., App. Phys. Letters 31, 37 (1977).

(3) Burnham, R., App. Phys. Letters 30, 132 (1977).

(4) Schimitschek, E.J., Celto, J.E., and Trias, J.A., App. Phys. Letters 31, 608 (1977).

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(6) Eyber, G., Z. Physik Chem. 144A, 1 (1929).

(7) See for example: Hutchinson, B., Hance, R.L., Bernard, B.B., and Hoffbauer, M., J. Chem. Phys. 63, 3694 (1975) and References therein.

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(9) Callear, A.B., and Oldman, R.J., Trans. Far. Soc., 63 2888 (1967).

(10) Rhodes, G., Ph.D. Thesis, University of Utah, 1973.

Our experimental arrangement is shown in Figure B-1. The pump laser is a commercial excimer laser (Tachisto, Inc., Needham, MA) which was modified to utilize a 50 cm focal length suprasil quartz lens as the output coupler.(11) Typical performance characteristics were 30-40 mJ of energy, 20 nsec pulse length and focused intensity of nearly a gigawatt/cm^2. The output was monitored for shot to shot variations by a photodiode and the total energy by a energy meter (Scientech model 362). The focused laser beam entered a 30 cm long reaction cell with the maximum flux inside the cell approximately 4 cm from the suprasil entrance window. For this configuration, optimum pressures of $Fe(CO)_5$ were near 0.2 torr. The output from the iron pentacarbonyl was observed in the forward scattered direction by a photodiode with discrimination against KrF photons being accomplished through the use of a mirror as well as chemical and narrow bandpass filters. Spectrally resolved output is shown in Figure B-2 where lines at 360, 385, 395, 540, 558 and 563 nm were observed. Time resolved signals from the photodiodes provided a measure of the temporal behavior of the output pulse relative to the pump laser pulse and showed the output to be prompt (see Figure B-3). Using narrow bandpass filters, we were able to show that 60% of the observed photons were due to the 385 nm transition with most of the rest at 563 nm. Overall conversion efficiency for this configuration was near 0.02%. Clearly, transverse pumping should provide more optimum coupling of the pump laser to the lasing media.

The spectral and temporal characteristics of the observed laser output suggests that inversion is due to an excited molecular electronic state of $Fe(CO)_5$ or a photofragment produced in the primary absorption process, e.g., $Fe(CO)_4^*$. Any process involving chemical reaction or energy transfer is too slow to be important on the time scale observed for the output pulse. Further experimentation is needed to assess the mechanism involved. Also, repeated irradiation with KrF produces particulate formation ($Fe_2(CO)_9$) and eventual reduction in output. It may be possible to use CO as a buffer gas and thereby extend the number of shots available from a single fill. A number of other metal-chelates exhibit similar absorption characteristics and could lead to similar lasing action due to photodissociation by high power excimer lasers.

The authors gratefully acknowledge Laboratory support provided by Mr. Anthony Montagna and Mr. Dilbert Yee.

(11) Barker, D. L., and Loree, T. R., App. Optics 16, 1792 (1977).

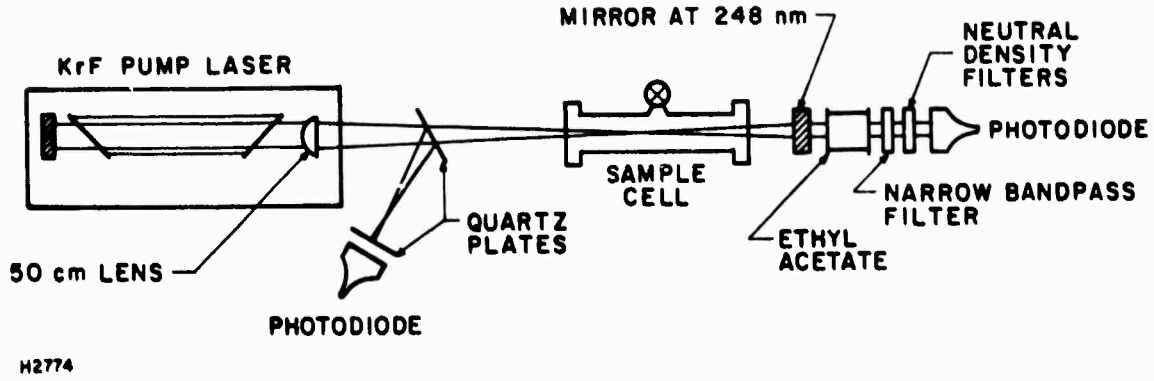

Figure B-1 Experimental Approach

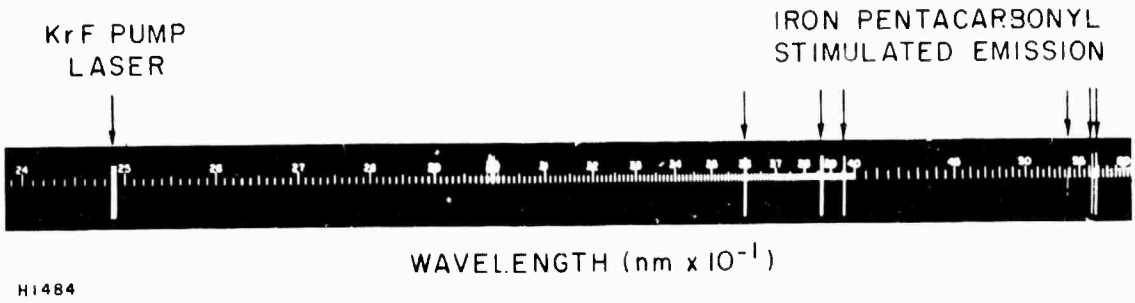

Figure B-2 Optically Pumped Iron Pentacarbonyl Lasing Transitions

OPTICALLY PUMPED IRON PENTACARBONYL
LASER OUTPUT

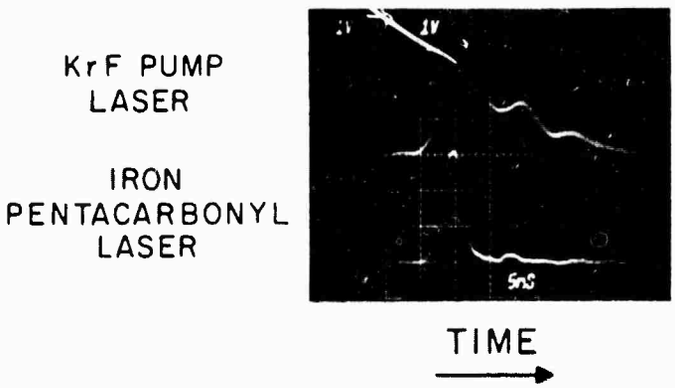

Figure B-3 Experimental Data

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